Summary

Cloud droplets and ice particles **play a pivotal role in modulating the Earth's climate through altering — sometimes drastically — radiative balance and precipitation.**(1; 2; 3) Nowadays, when climate change makes the headlines of the news on a daily basis, it is of utmost importance that we understand and characterize all physical processes which contribute to the complex mechanisms which determine the climate, in order to provide reliable predictions. Cloud droplet and ice nucleation are among the most complicated processes which define aerosol-cloud interactions. The lack of general understanding of heterogeneous nucleation, i.e. the process when droplet or ice condensation happens with catalytic effect of an insoluble surface (cloud or ice nucleating particle)(4) — in spite of the huge experimental and modelling efforts — is one main reason why aerosol-cloud interactions are to date poorly characterized in climate models.(5; 6) Challenges are multiple: 1)**The sheer chemical variety of droplet and ice nucleating surfaces**(7) can be by itself prohibitive for characterizing heterogeneous nucleation on a generalized level. 2) **The short time and length scales involved** further complicate the question, to the point that currently no experimental technique is able to observe nucleation events in real time.(8) 3) Finally, **not having a unified theory of heterogeneous nucleation** represents also a large obstacle. No wonder thus, that aerosol cloud interactions and ice parametrizations are not well constrained in climate models(6), which may lead to erroneous or contradictory predictions, whose correction has never been so urgent as today.

DIMolD aspires to remedy the issues which prevent the understanding of heterogeneous nucleation using a combination of observations and modeling to provide a **unified framework**, which can be **easily implemented** in climate models and **accounts for surface properties**, relying on a **theory which is closer to reality** than CNT and can be **informed from easily accessible data**. To achieve such converged theory one must exploit the spatio-temporal resolution of molecular simulations, which however must be validated against experiments. All this must be done keeping in mind that brute force efforts, both from the experimental and the computational side, to obtain data for CNT based parametrizations, despite the stellar computational cost of the latter, have not succeeded to date. Here thus I propose a dramatically different approach, I will use **inexpensive computational methods**, such as Grand Canonical Monte Carlo or equilibrium molecular dynamics simulations, and **standard CCN activity and contact angle measurements** as experimental techniques, abandoning the quest for the holy grail of obtaining nucleation rates, central for CNT based description of heterogeneous nucleation, for using **simple methods together with advanced thermodynamic description of the nucleation process to provide a better and more generalizable model for droplet and ice nucleation**. This approach is so far unprecedented and represents a radically changed viewpoint with respect to earlier studies in the field. This work may provide the missing link between simulations and measurements of heterogeneous nucleation, and more importantly it can be a great leap forward in producing unified theory of heterogeneous nucleation, which might be the way to push forward the limits of understanding this intriguing process and facilitate its implementation in climate models.